Fermilab
ES&H Section

RADIATION PHYSICS NOTE 136

Estimation of Leaching Parameters from NuMI Rock Boring Sample Data

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I. Introduction.

The primary driving force behind collecting the data presented in references 1 and 2 was to generate an empirical base which would allow the estimation of critical leaching parameters for the Fermilab ground water model (refs. 3,4 & 5) based on actual activated rock samples from the Fermilab site. The parameters of specific interest are the number of atoms of a given radionuclide produced per inelastic collision in a material (such as rock or soil), K_i, and the percentage of that radionuclide that is leached into ground water from the material matrix, L_i. For gamma emitting radionuclides both K_i and the product K_iL_i can be experimentally determined. However for sole beta emitters, such as ^3H, only the product K_iL_i can be experimentally determined, without invoking an experimental setup involving the destruction of one of two identical samples to establish the total ^3H generated by the irradiation.

II. ^{22}Na Analysis.

First, one needs to know the approximate total beam incident on the rock samples that produced the measured activation. Fortunately, the aluminum tags that were exposed with each rock sample provide enough information to estimate the beam flux. Then by scaling the experimental beam fluxes estimated from ^{22}Na activation in the aluminum tags by the ratio of cross sectional areas of the rock samples and the tags, i.e., 1.8013 ± 0.1037, one can estimate the number of particles incident on the rock samples. As in ref. 2, the ^{22}Na production cross section at a mean energy of 1 GeV was used to estimate the beam flux incident on the tags. Results from this exercise are presented in Table 1.

Next, the number of ^{22}Na atoms extant in the rock sample after radiation was calculated from the measured specific activity listed in column 2 of Table 2. Specific activity was converted to total activity by multiplying by the mass of each respective rock sample. This total activity was converted from picoCuries (pCi) to Bequerels (Bq) using the conversion factor of 27.03 pCi/Bq. One can then use the radioactive decay equation,

$$\frac{dN}{dt} = -\lambda N, \qquad (1)$$

Where $\frac{dN}{dt}$ is the observed decay rate in Bq and λ is the decay constant for ^{22}Na, to determine the number, N, of ^{22}Na atoms present in the rock matrix at termination of the irradiation. Table 2 presents N for each sample in column 5.

In order to estimate the number of inelastic interactions in each rock core sample during the irradiation period, one needs to evaluate the approximate inelastic cross sections for each sample. Since the rock core samples are known to be varying combinations of different minerals (ref. 6), primarily dolemite and shale, calculating an effective nuclear cross section for the mean molecular structure can be somewhat problematic. However, P. Kesich of the Fermilab ES&H Section's Environmental Protection group was kind enough to provide approximate estimates for percentage compositions of each rock core sample (ref. 7). Those estimates are presented in Table 3 along with the effective nuclear inelastic interaction cross section derived from them.

Effective nuclear inelastic cross sections for each molecular type, i.e., dolemite, shale, calcite, and pyrite were estimated by summing the nuclear inelastic cross section of each elemental constituent weighted by the number of atoms in each molecule. The elemental cross sections were obtained from reference 8, and the molecular formulas used were $CaMg(CO_3)_2$ for dolemite, $Al_2Si_2O_5(OH)_4$ for shale, $CaCO_3$ for calcite, and FeS_2 for pyrite. For example, the nuclear inelastic cross section for dolemite was estimated to be:

$$619 \quad + 396 + 2\times(231) + 6\times(292) = 3229\ mb$$

where 619 mb is the nuclear inelastic cross section for elemental calcium, 396 mb is the nuclear inelastic cross section for elemental magnesium, 231 mb is the nuclear inelastic cross section for elemental carbon, and 292 mb is the nuclear inelastic cross section for elemental oxygen. Nuclear inelastic cross sections for calcium and magnesium are not directly listed in ref. 8 and hence were obtained by a linear interpolation between adjacent elements listed in ref. 8. One can then estimate the nuclear inelastic cross sections for shale, calcite, and pyrite in analogous fashion as 4482 mb, 1726 mb, and 2126 mb respectively.

The effective nuclear inelastic cross section for each rock mixture represented by the rock core boring samples was then estimated by weighting each molecular constituent by its percent composition. For example, a mean nuclear inelastic cross section for sample #970318GL07 (row 4 in Table 3) is estimated as:

$$(0.70)\times(3229\ mb)+(0.20)\times(4482\ mb)+(0.025)\times(1726\ mb)+(0.075)\times(2126\ mb)=3359\ mb$$

It should be noted that there is approximately a 17% variation among the estimated cross sections. In a completely analogous manner, the mean GMW of the composite rock sample can also be estimated.

The number of inelastic nuclear interactions (stars) in each rock core sample can be precisely estimated using the individual molecular cross sections and gram molecular weights (GMWs) from preceding paragraphs and the formula;

$$I_T = \rho_a\ F_I\ A_V \sum_i w_i \frac{\sigma_i}{(GMW)_i} \qquad (2)$$

where:

- I_T ≡ Total number of inelastic interactions in a sample
- ρ_a ≡ Areal density for each respective rock core sample (grams/cm^2)
- σ_i ≡ Effective nuclear inelastic cross section for the ith molecule in the sample matrix. (cm^2-inelastic interactions /baryon-atom).
- A_v ≡ Avogrado's number of molecules per gram molecular weight. (atoms/GMW)
- F_I ≡ Total integrated baryon flux (baryons)
- w_i ≡ Fraction of rock matrix composed by ith molecular type.
- GMW_i ≡ Number of grams of ith molecule in a gram molecular weight. (grams/GMW).

Given the experimental error of 20% or greater, mean values for the cross section and GMW of the composite rock sample from Table 3 can be used in place of the summation in formula (2) with no noticeable change in the estimated values for K_i, L_i, and K_iL_i. Results are presented in Table 4 with the total number of inelastic interactions listed in column 7. The ratio of ^{22}Na leached out of the rock core samples to the total amount that existed in the rock sample at the start of the leaching process provides a good experimental estimate for L_i. This ratio can be calculated by multiplying the measured specific activity of the leachate (column 4 of Table 5) times the total volume of the leachate (column 2 of Table 5) divided by the product of the rock core mass (column 3 of Table 5) and the ^{22}Na specific activity in the sample at the beginning of the leaching process (column 5 of Table 5). Column 6 of Table 5 presents the final experimentally determined leaching ratio for ^{22}Na from the rock core samples tested.

Final experimentally determined parameters K_i and L_i for ^{22}Na in the NuMI rock core samples tested are presented in Table 6.

III. ^{3}H Analysis.

Results of the analysis for leachable ^{3}H in the NuMI rock core boring samples are summarized in Table 7. The recovered leachate volume and the ^{3}H specific activity in the leachate were directly measured. Their product, i.e., the total activity leached from the sample, is listed in the 4th column. The total number of leached ^{3}H atoms can then be calculated using the standard conversion factor between pCi and Bq and equation (1). The total number of inelastic interactions in each sample is the same as that listed in Table 4 and K_iL_i for ^{3}H can be calculated by dividing the total number of leached ^{3}H atoms by the total number of inelastic interactions (column 7).

IV. **Summary and Evaluation**.

It should be noted that, although most of the numbers in the Tables of this note are expressed to sometimes 6 decimal places to avoid rounding errors, final results have a precision of no more than 3 significant figures.

As a point of comparison, Table 10 of ref. 4 lists $K_i \approx 0.020$, Li ≈ 0.010, and $K_iL_i \approx 0.00020$ for ^{22}Na in dolemite. The experimentally determined values from this study were $K_i \approx 0.015$, Li ≈ 0.013, and the product $K_iL_i \approx 0.00019$. It should be noted that the experimentally determined numbers for L_i and K_iL_i both have a rather high random error of ~ 60% of the mean value. Random errors for the values from ref 4 were not listed so comparisons are only of marginal value, but for this type of measurement the agreement is unusually good.

Reference 4, Table 10 also lists $K_i \approx 0.030$, Li ≈ 1.000, and $K_iL_i \approx 0.030$ for ^{3}H in dolemite. The experimentally determined value from this study for K_iL_i is ≈ 0.00023 that is more than two orders of magnitude lower than the ref. 4 value. This discrepancy may be due to a drying out of the rock core samples before irradiation as was discussed in reference 2, and so must await future experimental determinations before any meaningful comparisons can be made.

V. **Acknowledgement**

The author would like to gratefully acknowledge the assistance provided by Alex Elwyn in reviewing and commenting on the initial draft of this note.

VI. **References**

1. V. Cupps, Residual Gamma Emitting Radionuclides in NuMI Rock Boring Samples, Fermilab Radiation Physics Note 133, February 1999.

2. V. Cupps, S. Benesch, and E. Kershisnik, Leachable ^{3}H and ^{22}Na in NuMI Rock Boring Samples, Fermilab Radiation Physics Note 134, December, 1999.

3. J.D. Cossairt, Use of a Concentration-Based Model for Calculating the Radioactivation of Soil and Groundwater at Fermilab, Fermilab Environmental Protection Note #8, Fermilab Environmental, Safety, and Health Section, 12/94.

4. A.J. Malensek, et. al., Groundwater Migration of Radionuclides at Fermilab, Fermilab-TM-1851, 8/93.

5. J.D. Cossairt, et. al., The Concentration Model Revisited, Fermilab Environmental Protection Note #17, June, 1999.

6. Hydrogeological Evaluation Report for the Fermilab Neutrino Main Injector (NuMI) Project, Volumes I &II, STS Consultants, Ltd., Deerfield, IL., 4/2/97.

7. P. Kesich, Fermilab ES&H Section Environmental Protection Group, private communication on 8/31/99.

8. Particle Data Group, *Atomic and Nuclear Properties of Materials Table*, p. 111.5, Review of Particle Properties, Physical Review D, Volume 45, #11, Part II, June 1992.

Table 1

Beam Flux incident of Rock Core Boring Samples at 1 GeV beam energy.

Fermilab Sample #	**Corresponding Tag #**	**Total Beam Estimated from Tags ($X10^{13}$)**	**Total Beam incident on rock disks ($X10^{13}$)**
970318GL02	5694	3.792	6.831
970318GL04	5692	5.067	9.127
970318GL06	5693	4.177	7.524
970318GL08	5695	3.823	6.886
970930GL01	5451	8.235	14.834
970930GL02	5453	6.144	11.067
970930GL03	5455	8.371	15.079
970930GL04	5454	10.06	18.121
970930GL05	5452	6.71	12.087
970930GL06	5456	7.141	12.863

Table 2

Number of ^{22}Na atoms generated in Rock Core Borings

Fermilab Sample #	**Measured ^{22}Na Concentration on reference date (pCi/gr)**	**Rock Core Mass (gms)**	**Total ^{22}Na Activity in Rock Core (Bq)**	**^{22}Na atoms at end of irradiation period**	**^{22}Na atoms per Baryon**
970318GL01	111.4	31.2692	128.871	1.526604E+10	2.235E-04
970318GL03	132.6	41.2553	202.384	2.397440E+10	2.627E-04
970318GL05	124.7	31.3247	144.513	1.711898E+10	2.275E-04
970318GL07	105.2	39.1939	152.542	1.807003E+10	2.624E-04
970930GL01	209.4	64.8703	502.547	5.953154E+10	4.014E-04
970930GL02	157.4	59.3662	345.699	4.095138E+10	3.699E-04
970930GL03	260.3	59.0668	568.816	6.738173E+10	4.468E-04
970930GL04	224.6	52.6804	437.737	5.185413E+10	2.862E-04
970930GL05	178.5	65.4403	432.153	5.119270E+10	4.234E-04
970930GL06	195.7	49.4563	358.069	4.241671E+10	3.298E-04

Table 3

Approximate Composition and Mean Nuclear Inelastic Cross Sections of NuMI Rock Core Boring Samples

Fermilab Sample #	Depth of Core Sample (ft)	Percent Dolemite	Percent Shale	Percent Calcite	Percent Pyrite	Mean GMW (grams)	Mean Nuclear Inelastic Cross Section (mb)
970318GL01	185	70.00	30.00			206.5343	3605
970318GL03	130	50.00	50.00			221.2849	3856
970318GL05	138	50.00	50.00			221.2849	3856
970318GL07	107	70.00	20.00	2.50	7.50	192.1504	3359
970930GL01	124.5	80.00	15.00	2.50	2.50	191.7298	3352
970930GL02	200	50.00	45.00	2.50	2.50	213.8557	3728
970930GL03	157.5	85.00	10.00	2.50	2.50	188.0422	3289
970930GL04	125	80.00	15.00	2.50	2.50	191.7298	3352
970930GL05	153	65.00	30.00	2.50	2.50	202.7928	3540
970930GL06	200	50.00	45.00	2.50	2.50	213.8557	3728

Table 4

Number of Inelastic Interactions generated in the Rock Core Samples

Fermilab Sample #	Depth of Core Sample (ft)	Rock Core Thickness (cm)	Areal Density (gms/cm^2)	Beam Incident on sample (baryons) (X10^{13})	Effective Nuclear Inelastic X-Section (mb)	Mean GMW (grams)	Inelastic interactions in Sample (X 10^{12})
970318GL01	185	0.65	1.781	6.831	3605	206.5343	1.278798115
970318GL03	130	0.7	1.918	9.127	3856	221.2849	1.836972148
970318GL05	138	0.65	1.781	7.524	3856	221.2849	1.406172579
970318GL07	107	0.65	1.781	6.886	3359	192.1504	1.291042382
970930GL01	124.5	0.85	2.329	14.834	3352	191.7298	3.637331262
970930GL02	200	0.8	2.192	11.067	3728	213.8557	2.546631994
970930GL03	157.5	0.7	1.918	15.079	3289	188.0422	3.046284909
970930GL04	125	0.7	1.918	18.121	3352	191.7298	3.65919754
970930GL05	153	0.8	2.192	12.087	3540	202.7928	2.785162812
970930GL06	200	0.75	2.055	12.863	3728	213.8557	2.774915911

Table 5

Proportion of ^{22}Na atoms leached from the NuMI rock core samples

Fermilab Sample #	Recovered Leachate Volume (ml)	Rock Core Mass (gms)	^{22}Na Specific Activity in Leachate (pCi/ml)	^{22}Na Specific Activity in Sample at start of Leaching (pCi/gms)	Ratio of ^{22}Na leached from sample (L_i)
970318GL01	112	31.2692	0.71	107.00	0.023767
970318GL03	116	41.2553	0.41	127.00	0.009077
970318GL05	114	31.3247	0.51	120.00	0.015467
970318GL07	116	39.1939	0.31	101.00	0.009084
970930GL01	99	64.8703	1.00	187.00	0.008161
970930GL02	94	59.3662	2.60	140.00	0.029406
970930GL03	48	59.0668	0.88	200.00	0.003576
970930GL04	94	52.6804	1.03	232.00	0.007922
970930GL05	52	65.4403	1.67	159.00	0.008346
970930GL06	41	49.4563	3.15	175.00	0.014922

Table 6

Final Experimental Parameters K_iL_i for ^{22}Na NuMI rock core Samples

Fermilab Sample #	**Inelastic Interactions in Sample ($X10^{12}$)**	**# of ^{22}Na atoms produced in Sample ($X10^{10}$)**	**# of ^{22}Na atoms produced per inelastic interaction (K_i)**	**Ratio of ^{22}Na leached from sample (L_i)**	**K_iL_i**
970318GL01	1.278798115	1.5266	0.011938	0.023767	0.00028373
970318GL03	1.836972148	2.39744	0.013051	0.009077	0.00011846
970318GL05	1.406172579	1.711898	0.012174	0.015467	0.00018830
970318GL07	1.291042382	1.807003	0.013996	0.009084	0.00012714
970930GL01	3.637331262	5.953154	0.016367	0.008161	0.00013357
970930GL02	2.546631994	4.095138	0.016081	0.029406	0.00047287
970930GL03	3.046284909	6.738173	0.022119	0.003576	0.00007910
970930GL04	3.65919754	5.185413	0.014171	0.007922	0.00011226
970930GL05	2.785162812	5.11927	0.018381	0.008346	0.00015340
970930GL06	2.774915911	4.241671	0.015286	0.014922	0.00022809

Average =	0.015356335	0.0129728	0.000189693
Random Error =	0.003106	0.008064024	0.000116517
% Error	20%	62%	61%

Table 7

Number of ^3H atoms leached from NuMI rock core samples

Fermilab Sample #	Recovered Leachate Volume (ml)	^3H Specific Activity in Leachate (pCi/ml)	Total ^3H Activity Leached from Sample (pCi)	Total # of leached ^3H atoms	Inelastic Interactions in Sample ($X10^{12}$)	K_iL_i for ^3H
970318GL01	112	0.15	16.80	3.48894E+08	1.278798	2.72829E-04
970318GL03	116	0.12	13.92	2.89083E+08	1.836972	1.57369E-04
970318GL05	114	< 0.10	< 11.4	< 2.36749E+08	1.406173	< 1.40307E-04
970318GL07	116	< 0.10	< 11.6	< 2.40903E+08	1.291042	< 1.79078E-04
970930GL01	99	0.33	32.67	6.78473E+08	3.637331	1.86531E-04
970930GL02	94	0.73	68.62	1.42506E+09	2.546632	5.59588E-04
970930GL03	48	0.68	32.64	6.77850E+08	3.046285	2.22517E-04
970930GL04	94	0.16	15.04	3.12343E+08	3.659198	8.53583E-05
970930GL05	52	0.52	27.04	5.61553E+08	2.785163	2.01623E-04
970930GL06	41	0.53	21.73	4.51277E+08	2.774916	1.62627E-04

K_iL_i Average =	2.31055E-04
Error =	1.4340E-04
%Error =	62%